

**Amendments to the Specification:**

**Please replace the paragraph beginning on page 1, line 15 and ending on page 2, line 2, with the following amended paragraph:**

The ~~structure of~~ semiconductor light-emitting diode (LED) structure comprises a substrate, a light emitting structure, and a pair of electrode for powering the diode. The substrate can be an opaque or a transparent substrate. The Light emitting diodes ~~[[that]]~~ are based on gallium nitride compounds which generally comprise: a transparent ~~[[,]]~~ and insulating substrate, e.g., a sapphire substrate. In ~~generally general~~, ~~[[To]]~~ to overcome ~~the difficulty of~~ the substantial lattice mismatch between ~~[[an]]~~ the insulating substrate, e.g., a sapphire substrate, and GaN compound semiconductor, it is a common practice to provide a thin buffer layer or a nucleation layer on the ~~sapphire~~ insulating substrate, which is formed followed by a layer on which an LED structure is grown. The ~~Growth~~ growth of single crystals on the insulating substrates that has been studied for many years. Early works included the growth of both silicon and III-V compound semiconductors on a variety of insulating substrates that including sapphire. In these studies, it was determined ~~that use the~~ usage of nucleation or buffer layers is to ~~reduces~~ reduce the occurrence of imperfections and the tendency towards twinning in the thicker layer grown thereon.

**Please replace the paragraph on page 2, lines 4-21, with the following amended paragraph:**

~~Group~~ Group-III nitride semiconductors [GaN (gallium nitride), InN (indium nitride), AlN (aluminum nitride), and their alloys] have become the materials of choice for many optoelectronic applications, especially in the areas of fully-color or ~~white-light~~ white light-emitting diodes (LEDs) and blue laser diodes (LDs). Some scientists and engineers have even predicated that group-III ~~nitrides~~ nitride

semiconductors will become all-around semiconductors besides their already-commercialized applications in optoelectronics. At present, the major barrier for widespread applications of nitrides is ~~[[the]]~~ lack of perfectly lattice-matched substrates for epitaxial growth. Sapphire ( $\text{Al}_2\text{O}_3$ ) and silicon carbide (SiC) are two most popular materials ~~choices~~ as growth the substrates. ~~[[But,]]~~ Beside the large lattice mismatch, the ~~insulating nature~~ insulation property of sapphire renders the processing of nitride devices more difficult and costly. On the other hand, the high price and limited size of silicon carbide also make the widespread GaN-on-SiC applications difficult. GaN-on-Si epitaxial technology represents an interesting alternative, which can eventually integrate the existing Si-based microelectronic technology and the novel functionalities provided by the group-III nitrides.

**Please replace the paragraph beginning on page 2, line 23 and ending on page 3, line2, with the following amended paragraph:**

For GaN-on-Si heteroepitaxy, the AlN single-layered buffer layer approach yields can provide the best good results as reported in the literature, and leading to the demonstration of light-emitting diodes on Si. However, the mutual solubility of Al and Si is very high at the AlN buffer-layer growth temperature (about 820°C vs. eutectic temperature 577°C). Therefore, the inter-diffusion of Al and Si at the interface is severe, resulting in high unintentional doping levels in the epilayer and the Si substrate as well as the degradation in the film structural and optical quality.

**Please replace the paragraph on page 3, lines 4-19, with the following amended paragraph:**

On the other hand, ~~[[It]]~~ it has been found that an amorphous or polycrystalline  $\text{SiN}_x$  [silicon nitride ( $\text{Si}_3\text{N}_4$ ) or silicon subnitride] layer can be formed by intentional or unintentional nitridation of the silicon substrate surface during the first stage of the group-III nitride growth. Moreover,  $\text{Si}_3\text{N}_4$  is well known to be an effective diffusion barrier material. However, this amorphous or polycrystalline  $\text{SiN}_x$  layer is prone to cause detrimental effects on the properties of ~~grown~~ GaN films grown on the Si substrate, since it is not possible to grow a high-quality crystalline film on ~~[[a]]~~ an amorphous or polycrystalline surface. Therefore, it has been a common practice in the growth of group-III-nitrides film on the silicon substrate to avoid the formation of an amorphous or polycrystalline  $\text{SiN}_x$  layer during the first stage of the group-III nitride growth. To overcome the effects of amorphous or polycrystalline  $\text{SiN}_x$  on the growth quality and to facilitate an effective diffusion barrier layer, the formation of a single-crystal diffusion barrier layer which can be lattice matched to the Si(111) surface is highly desirable.

**Please replace the paragraph on page 4, lines 2-5, with the following amended paragraph:**

It is a further object of this invention to provide a single-crystal  $\text{AlN/Si}_3\text{N}_4$  ~~double-buffer-layer~~ double-layered buffer with coincident lattice conditions on a Si(111) substrate that can alleviate the problems of lattice mismatch and interdiffusion, thereafter inducing high-quality heteroepitaxial growth.

**Please replace the paragraph on page 4, lines 7-27, with the following amended paragraph:**

According to abovementioned objects, the present invention provides a structure for resolving the issue of auto-doping, resulting from Al/Si, Ga/Si, or  $[[\text{InN}]]$  In/Si inter-diffusion when grown with a group-III nitride buffer layer. The structure comprises a Si(111) substrate that surface has been reconstructed by *in-situ* thermal annealing to remove the remained thin oxide layer and to prepare clean and smooth silicon surface at high temperature. Then, the key feature of the present invention, a multiple-layered buffer layer is formed on the reconstructed Si(111) substrate. The multiple-layered buffer includes a single-crystal silicon nitride layer and a single-crystal AlN layer or other group-III nitride semiconductor epitaxial layer thereon. Next, the GaN epilayer is grown on the multiple-layered buffer. The advantage of the present invention is that the multiple-layered buffer ~~mechanism~~ can improve heteroepitaxial growth with large lattice mismatch. Furthermore, the 1:2 and 5:2 coincident lattices formed at the interface of the single-crystal silicon nitride ( $\text{Si}_3\text{N}_4$ )/Si(111) and the interface of the single-crystal aluminum nitride ~~AlN(0001)~~/single-crystal silicon nitride ( $\text{Si}_3\text{N}_4$ ) respectively can be used to facilitate the multiple-layered buffer for high-quality GaN-on-Si heteroepitaxial growth. Thus, the inter-diffusion between group-III elements (Al,  $[[\text{In}]]$  Ga, or  $[[\text{Ga}]]$  In) and Si can be resolved and the epitaxial growth quality can be improved.

**Please replace the paragraph on page 5, lines 1-11, with the following amended paragraph:**

Moreover, the present invention provides a method for forming a group-III heteroepitaxial structure on a Si(111) substrate. The key feature of the present invention is that the multiple-layered buffer is formed on the Si(111) substrate. The

multiple-layered buffer comprises a single-crystal silicon nitride ( $\text{Si}_3\text{N}_4$ ) layer that is formed on the Si(111) substrate by introducing reactive nitrogen-plasma to the reconstructed Si(111) surface in the controlled manner to prevent the formation of amorphous or polycrystalline  $\text{SiN}_x$  layer. Then, another buffer layer is an AlN layer or other group-III nitride layer, which is grown epitaxially on the single-crystal silicon nitride layer. Similarly, the group-III nitride heteroepitaxial structure is epitaxially grown on the AlN layer.

**Please replace the paragraph on page 6, lines 2-6, with the following amended paragraph:**

FIG. 3 is a schematic representation the SIMS depth profiles near the buffer/substrate and the epilayer/buffer interface regions for samples with a single-crystal AlN/ $\text{Si}_3\text{N}_4$  ~~double-buffer-layer~~ double-layered buffer (a) and a single-crystal AlN single-layered buffer layer (b) in accordance with a method disclosed herein;

**Please replace the paragraph on page 6, lines 12-16, with the following amended paragraph:**

FIG. 4B is a schematic representation showing the Arrhenius plots of the luminescence intensities of free exciton (FX) grown on a single-crystal AlN/ $\text{Si}_3\text{N}_4$  ~~double-buffer-layer~~ double-layered buffer and neutral-donor-bound exciton grown on single-crystal AlN single buffer layer in accordance with a structure disclosed herein; and

**Please replace the paragraph beginning on page 7, line 22 and ending on page 8, line 7, with the following amended paragraph:**

The stacked buffer ~~[[layer]]~~ consists of constituent layers, which can form coincident lattices at layer/layer and layer/substrate interfaces. In the preferred embodiment of the present invention, the buffer ~~[[layer]] comprising~~ comprises at least two layers of distinct material with sharp material transitions and epitaxial alignments between the layers and between the bottom layer of the buffer layer and the Si(111) substrate. For the case of GaN-on-Si(111) heteroepitaxy, the present invention utilizes the 1:2 and 5:2 coincident lattices formed at the  $\beta$ -Si<sub>3</sub>N<sub>4</sub> ~~(0001)~~ {0001}/Si(111) and AlN ~~(0001)~~ {0001}/ $\beta$ -Si<sub>3</sub>N<sub>4</sub>~~(0001)~~ {0001} interfaces respectively to facilitate the ~~double-buffer layers~~ double-layered buffer for high-quality GaN-on-Si heteroepitaxial growth. By using this buffer technique, the present invention can resolve the issue of auto-doping, resulting from Al/Si inter-diffusion when grown with a single AlN ~~(0001)~~ {0001} coincident buffer. As the result, the epitaxial quality of GaN film is also significantly improved.

**Please replace the paragraph on page 8, lines 9-17, with the following amended paragraph:**

GaN on Si(111) heteroepitaxy constitutes a large +20.4% in-plane lattice mismatch ( $\equiv (a_{\text{Si}} - a_{\text{GaN}})/a_{\text{GaN}}$ ;  $a_{\text{GaN}(\text{0001})\text{0001}} = 3.189\text{\AA}$ ;  $a_{\text{Si}(111)} = 3.840\text{\AA}$ ) and large thermal expansion mismatch. Fortunately, by using a buffer layer with coincident lattice conditions can alleviate the lattice mismatch. For example, a 5:4 lattice coincidence between AlN ~~(0001)~~ {0001} ( $a_{\text{AlN}(0001)} = 3.112\text{\AA}$ ) and Si(111) can reduce the lattice mismatch from 23.4% (tensile strain) to an effective lattice mismatch of -1.3% (compressive strain). As a result, two-dimensional smooth epitaxial growth mode was found to be possible due to the reduced strain.

**Please replace the paragraph beginning on page 8, line 19 and ending on page 9, line 6, with the following amended paragraph:**

The growth processes were conducted in a molecular-beam epitaxy (MBE) apparatus equipped with a radio frequency (RF) nitrogen plasma source. The base pressure in the MBE growth chamber ~~[[was]]~~ is about  $6 \times 10^{-11}$  ~~[[Torr]]~~ torr. High-purity Ga and Al metals ~~[[were]]~~ are used for the conventional effusion cells. Nitrogen gas (N<sub>2</sub>) ~~[[was]]~~ is purified through a nitrogen purifier before fed into the radio-frequency ~~[[rf]]~~ plasma source. Nitrogen plasma ~~[[was]]~~ is generated under the same conditions during all the growth processes. The RF power ~~[[is]]~~ was about 450 watt and the nitrogen flow rate of the nitrogen ~~[[is]]~~ was about 0.5 sccm. Three-inch Si(111) substrate (boron-doped p-type) ~~[[was]]~~ is chemical etched before loading into the MBE chamber. The Si(111) substrate ~~[[was]]~~ is further thermally annealed *in situ* to remove the remained thin oxide layer and to prepare a clean and smooth silicon surface at high temperature. The Si(111) substrate prepared by this process showed a clear ~~(7\*7)~~ (7x7) surface reconstruction, confirmed by the reflection high-energy electron diffraction (RHEED) pattern at about 800°C.

**Please replace the paragraph on page 9, lines 8-27, with the following amended paragraph:**

Furthermore, the RHEED pattern indicates a high-quality and smooth reconstructed Si surface prior to the growth process. The substrate temperature was calibrated by observing the ~~(7\*7)~~ (7x7) to ~~(1\*1)~~ (1x1) phase transition of Si(111) surface at 875°C. In the present invention, two different buffer-layer systems for GaN growth on Si (111) substrates were prepared for comparison. Both samples consist of an AlN buffer layer with a thickness of 30 nm. The only difference is that one sample

contains a single-crystal  $\beta$ -Si<sub>3</sub>N<sub>4</sub> layer [1.5-nm-thick layer with an abrupt interface with the Si(111) substrate, as confirmed by transmission electron microscopy (TEM)] before the AlN layer growth. Single-crystal silicon nitride layer can be formed by nitridation of the Si(111) surface under the reactive nitrogen plasma for 30 ~~[[sec]]~~ seconds at a substrate temperature of about 900°C. The 30 nm thick AlN buffer layers were grown epitaxially at 820°C with a growth rate of 0.12  $\mu\text{m/hr}$ . Furthermore, the 240-nm-thick GaN epitaxial layers ~~[[were]]~~ are grown on the buffer layers at a lower substrate temperature (720°C) with a growth rate of 0.08 ~~[[ $\mu\text{m/hr}$ ]]~~  $\mu\text{m/hr}$ . ~~After the MBE growth, the grown GaN surface (cooling to 500 to 600°C) showed a (2\*2) reconstruction pattern under the nitrogen flux, indicating a Ga polar film. The Ga polar GaN film is known to have better structural and optic properties~~

**Please replace the paragraph on page 10, lines 1-25, with the following amended paragraph:**

FIG. 1 shows a flow chart of the method for forming a ~~double-buffer-layer~~ double-layered buffer on the Si(111) substrate, wherein, FIG. 1 divided into FIG. 1A and FIG. 1B to show the formation process of ~~double-buffer-layer~~ double-layered buffer on the Si(111) substrate. Step 1 illustrates the Si(111) substrate that were thermally annealed *in situ* to remove the remained thin oxide layer and to prepare a clean and smooth silicon surface at high temperature. The Si(111) substrate prepared by the process showed a clear ~~(7\*7)~~ (7x7) surface reconstruction, that can be confirmed by the RHEED pattern at about 800°C. Step 2 illustrates the reactive nitrogen-plasma is introduced to the surface of the reconstructed Si (111) substrate in a controlled manner to form the single-crystal silicon nitride (Si<sub>3</sub>N<sub>4</sub>) diffusion-barrier buffer layer by nitridation of the surface of the Si (111) substrate. The surface of single-crystal Si<sub>3</sub>N<sub>4</sub> diffusion-barrier buffer layer formed in step 2 is terminated by nitrogen surface adatoms. Step 3 illustrates the process for forming Al pre-deposition



atomic layer on the single-crystal nitrogen-terminated  $\text{Si}_3\text{N}_4$  diffusion-barrier buffer layer. Then, a thermal annealing process is performed to the Al pre-deposition atomic layer to form an AlN monolayer on the single-crystal  $\text{Si}_3\text{N}_4$  diffusion-barrier buffer layer without the reactive nitrogen species (step 4). Next, an epitaxial AlN buffer layer is formed on the single-crystal  $\text{Si}_3\text{N}_4$  diffusion-barrier buffer layer by performing an AlN epitaxial growth process on the AlN monolayer (step 5). Finally, ~~[[the]]~~ a GaN epitaxial film with ~~a Ga-polarized surface~~ or a group-III nitride semiconductor heteroepitaxial structure is grown by an epitaxial growth method on the single-crystal AlN buffer layer (step 6).

**Please replace the paragraph beginning on page 10, line 27 and ending on page 11, line 9, with the following amended paragraph:**

Then, referring to FIG. 2A, the Si(111) substrate 10 is initially treated by *in-situ* annealing process or *ex-situ* wet etching process (such as etching by an HF solution) to remove the remained thin oxide layer and to prepare a clean and smooth silicon surface. The Si(111) substrate 10 prepared by the thermal annealing process shows a clear ~~(7\*7)~~ (7x7) surface reconstruction, and is confirmed by the reflection high-energy electron diffraction (RHEED) pattern at about 800°C. The RHEED pattern can be used to ~~indicate~~ confirm a high-quality and smooth reconstructed Si(111) substrate surface prior to the growth processes. The substrate temperature is calibrated by observing the ~~(7\*7)~~ (7x7) to ~~(1\*1)~~ (1x1) phase transition of Si(111) surface at 875°C.

**Please replace the paragraph on page 11, lines 11-25, with the following amended paragraph:**

Next, the key feature of the present invention is that the diffusion-barrier buffer layer 12 such as a single-crystal silicon nitride layer is formed by nitrogen-plasma nitridation of the surface of Si(111) substrate 10. The nitridation process is performed by exposing the surface of Si(111) substrate 10 to the reactive nitrogen plasma for about 30 seconds at a substrate temperature of about 900°C. Exposure time is critically controlled to prevent the unwanted formation of amorphous or polycrystalline SiN<sub>x</sub> layer. In the present invention, the single-crystal silicon nitride [ $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0001~~) {0001}] layer 12 can also be formed on the Si(111) substrate 10 by introduction of reactive nitrogen-containing species including NH<sub>3</sub> while the surface of Si (111) substrate 10 held slightly higher than the (~~7\*7~~) (7x7) to (~~1\*1~~) (1x1) phase transition temperature. A single-crystal (~~4\*4~~) (4x4) surface-reconstruction (alternatively, the “(~~8\*8~~) (8x8)”-reconstruction in terms of the Si (111) substrate 10 lattice parameter) usually forms after such controlled nitridation process.

**Please replace the paragraph beginning on page 11, line 27 and ending on page 12, line 7, with the following amended paragraph:**

The RHEED pattern shows the “(~~8\*8~~) (8x8)”-reconstructed surface after introducing the reactive nitrogen plasma to the surface of Si(111) substrate at the substrate temperature of 900°C for about 30 seconds. The RHEED pattern shows that are two different ordering on  $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0001~~) {0001}. One ordering corresponding to the topmost “(~~8/3\*8/3~~) (8/3x8/3)”-ordered nitrogen adatoms and the other corresponds to the “(~~8\*8~~) (8x8)” lattice periodicity. In the previous scanning tunneling microscopy experiments, the “(~~8\*8~~) (8x8)” ordering was confirmed to be the

reconstruction unit cell of the  $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0001~~) {0001} surface.

**Please replace the paragraph on page 12, lines 9-26, with the following amended paragraph:**

Then, the growth of AlN buffer layer 16 in the ~~double-buffer-layer~~ double-layered buffer system was started on the nitrogen-terminated Si<sub>3</sub>N<sub>4</sub> reconstruction surface as shown in FIGS. 2B and 2C. After 15 seconds of Al pre-deposition process, the Al atomic layer 14 is formed on the surface of single-crystal Si<sub>3</sub>N<sub>4</sub> layer 12. Then, a single-crystal AlN buffer layer 16 stacked on the single-crystal Si<sub>3</sub>N<sub>4</sub> layer 12 is formed first by performing a thermal annealing process to the AlN pre-deposition atomic layer 14. The AlN (0001)-(~~1\*1~~) (1x1) ordering appears in the streaky RHEED pattern after the thermal annealing step. The RHEED pattern indicates that Al atoms are bounded with the topmost N adatoms of the single-crystal Si<sub>3</sub>N<sub>4</sub> layer 12 and the surface is very smooth. It should be noted that the reciprocal space periodicities along the bulk  $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0-110~~) <2-1-10> 12 and AlN(~~0-110~~) <2-1-10> 16 directions are 4:5 and this condition can be confirmed in the RHEED pattern. Then, an AlN buffer layer 16 is grown by an epitaxial growth method on the single-crystal Si<sub>3</sub>N<sub>4</sub> layer 12. In addition, it is also possible that the second buffer layer stacked on the single-crystal Si<sub>3</sub>N<sub>4</sub> diffusion-barrier buffer layer 12 is a GaN (gallium nitride) layer or InN (indium nitride) layer.

**Please replace the paragraph beginning on page 12, line 28 and ending on page 13, line 10, with the following amended paragraph:**

In order to compare the effects of single-crystal AlN single-layered buffer and AlN/Si<sub>3</sub>N<sub>4</sub> ~~double-buffer-layer~~ double-layered buffer on the grown film quality, the

AlN buffer layer 16 with an identical thickness about 30 nm were grown epitaxially on two Si substrates with and without the single-crystal Si<sub>3</sub>N<sub>4</sub> layer 12 at a substrate temperature of 820°C ~~[[with]]~~ and a growth rate of 0.12 ~~[[um/hr]]~~ μm/hr, ~~as shown in FIG. 2D.~~ Moreover, the GaN epitaxial layers 20 with an identical thickness about 240 nm were grown on the single-crystal AlN buffer layers 16 of these two samples at a lower substrate temperature which is about 720°C with a growth rate of 0.08 ~~[[um/hr]]~~ μm/hr. ~~After the MBE growth, the grown GaN surface after cooling to 500 to 600°~~ shows a (2\*2) ~~reconstruction pattern under the nitrogen plasma flux.~~

**Please replace the paragraph on page 13, lines 12-24, with the following amended paragraph:**

As referring to FIG. 2D, the present invention provides a light-emitting device structure with a ~~double buffer layer~~ double-layered buffer to resolve the issue of the auto-doping, resulted from the inter-diffusion of Al/Si and Ga/Si when grown with a single AlN(~~0001~~) {0001} coincident buffer. The light-emitting device provides a Si(111) substrate 10 with in-plane lattice constant of 3.84 angstroms. The key feature of the present invention is a ~~double buffer layer~~ double-layered buffer on the Si(111) substrate 10. In the preferred embodiment of the present invention, the ~~double buffer layer~~ double-layered buffer can improve the light-emitting efficiency for the light emitting device, wherein the ~~double buffer layer~~ double-layered buffer includes a single crystal silicon nitride layer (Si<sub>3</sub>N<sub>4</sub>) (~~0001~~) {0001} 12 with in-plane lattice constant 7.61 angstroms, and the AlN(~~00001~~) {0001} layer 16 with in-plane lattice constant 3.112 angstroms.

**Please replace the paragraph beginning on page 13, line 26 and ending on page 14, line 12, with the following amended paragraph:**

In addition, the present invention shows a streaky RHEED pattern after 10-min AlN growth at 840°C and indicates that the resulting epitaxial AlN buffer layer 16 has a smooth surface and is of high film quality. A GaN epilayer grown on the single-crystal Si<sub>3</sub>N<sub>4</sub>/AlN ~~double-buffer-layer~~ double-layered buffer by MBE also has a smooth surface morphology and high crystalline quality as demonstrated by the *in-situ* streaky RHEED pattern and is confirmed by *ex-situ* X-ray diffraction (XRD) and atomic force microscopy (AFM) measurements. Herein, the possible overgrown heteroepitaxial structure on top of the ~~double-buffer-layer~~ double-layered buffer includes a group-III nitride semiconductor single epitaxial layer or group-III nitride semiconductor ~~heteroepitaxial multiple-layer~~ multiple heteroepitaxial layers. From the *in-situ* RHEED and *ex-situ* XRD measurements, the present invention can determine that 1:2 and 5:2 coincident lattice interfaces are formed at  $\beta$ -Si<sub>3</sub>N<sub>4</sub>(0001) ~~{0001}~~/Si(111) and AlN(0001)/ $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0001~~) ~~{0001}~~ interfaces, respectively.

**Please replace the paragraph on page 14, lines 14-25, with the following amended paragraph:**

Furthermore, the following epitaxial orientation relationships are found by the RHEED and XRD studies:  $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0001~~) ~~{0001}~~||Si(111);  $\beta$ -Si<sub>3</sub>N<sub>4</sub>[~~0 $\bar{1}$ 10~~] ~~<-1 1 0>~~||Si[11 $\bar{1}$ ][ $\bar{2}$ ] ~~-2~~];  $\beta$ -Si<sub>3</sub>N<sub>4</sub>[~~2 $\bar{1}$  $\bar{1}$ 0~~] ~~<2 -1 -1 0>~~||Si[ $\bar{1}$ 10] ~~<-1 1 0>~~ and AlN(~~0001~~) ~~{0001}~~|| $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0001~~) ~~{0001}~~; AlN[~~0 $\bar{1}$ 10~~] ~~<0 -1 1 0>~~|| $\beta$ -Si<sub>3</sub>N<sub>4</sub>[~~0 $\bar{1}$ 10~~] ~~<0 -1 1 0>~~; AlN[~~2 $\bar{1}$  $\bar{1}$ 0~~] ~~<2 -1 -1 0>~~|| $\beta$ -Si<sub>3</sub>N<sub>4</sub>[~~2 $\bar{1}$  $\bar{1}$ 0~~] ~~<2 -1 -1 0>~~. Thus, the GaN/AlN/ $\beta$ -Si<sub>3</sub>N<sub>4</sub> *c*-axis is perpendicular to the surface of Si(111) substrate. It is tempting to perform heteroepitaxy of GaN on the Si(111) substrate using a single  $\beta$ -Si<sub>3</sub>N<sub>4</sub> buffer layer (without the AlN buffer layer). However, the present invention confirms that the

resulting growth is rough and polycrystalline in the first growth stage as indicated by the spotty RHEED pattern during the initial GaN film growth. Therefore, the ~~double buffer layer~~ double-layered buffer approach can yield better interface properties between the epilayer and the buffer ~~[[layer]]~~.

**Please replace the paragraph on page 15, lines 7-20, with the following amended paragraph:**

Firstly, the impurity distribution in the growth direction can be detected by SIMS. In order to investigate the auto-doping effects while GaN grown on the Si(111) substrate, the present invention focus on Al and Si ion signal depth profiles in the GaN/AlN and AlN/Si interface regions. The SIMS spectra were obtained by using a 7.7 keV Cs<sup>+</sup> primary beam and were used to probe Al and Si depth profiles in GaN films grown on Si(111) substrate using two buffer layer systems. The depth zero points of each SIMS spectrum are set at the top of Si(111) substrates. ~~[[Focus]]~~ Focusing on the Si depth profiles in AlN buffers and GaN epitaxial layers, the magnitudes of the Si ion signal are indicated by the solid arrows in FIG. 3. From the SIMS spectra, the magnitudes of Si impurities in the AlN buffer layer and the GaN film using a single-crystal  $\beta$ -Si<sub>3</sub>N<sub>4</sub>(~~0001~~) {0001}/AlN(~~0001~~) {0001}/Si(111). ~~double buffer layer~~ double-layered buffer is about one order of magnitude lower than that using an AlN single-layered buffer ~~[[layer]]~~.

**Please replace the paragraph beginning on page 15, line 22 and ending on page 16, line 2, with the following amended paragraph:**

Not only the single-crystal Si<sub>3</sub>N<sub>4</sub> layer inhibits the Si diffusion into the AlN and GaN layers, it also prevents the Al diffusion into the silicon substrate during the high

-temperature growth of AlN buffer layer. And, the magnitude of the Al ion signal in Si(111) substrate is also about one order of magnitude lower than that grown without the single-crystal ~~silicon-nitride~~  $\text{Si}_3\text{N}_4$  diffusion barrier layer. Therefore, the SIMS spectra show that single-crystal  $\text{Si}_3\text{N}_4$  layer effectively ~~inhibits~~ prevents the Si diffusion into the AlN and GaN layer and the Al diffusion into the silicon substrate ~~effectively~~ during the AlN high-temperature growth and the sequential GaN ~~epitaxial~~ growth stages.

**Please replace the paragraph on page 16, lines 4-22, with the following amended paragraph:**

FIGS. 4A and 4B show the comparison of the optical properties of GaN films that grown on different buffer layers. The low-temperature (6.7 K) PL spectra indicate that the GaN film grown on single-crystal AlN/ $\text{Si}_3\text{N}_4$ /Si(111) has a smaller full width at half maximum (FWHM) of neutral-donor-bound excitation ( $\text{D}^0\text{X}$ ) near-band-edge luminescence peak than that of the GaN film grown on AlN(0001)/Si(111). The decrease in the FWHM value of PL peak (12 meV vs. 20 meV) is consistent with the deduction of dislocation density ( $[[7*10^8]]$   $7 \times 10^8$   $\text{cm}^{-2}$  vs.  $[[1.1*10^9]]$   $1.1 \times 10^9$   $\text{cm}^{-2}$ ) measured by AFM, confirming a significant improvement in the epilayer crystalline quality. The inset in FIG. 4A displays the main luminescence peak position in the PL spectra of GaN grown on single-crystal AlN/ $\text{Si}_3\text{N}_4$ /Si(111) at different temperatures, indicating that the dominant PL peak changes to the free exciton (FX) emission at increasing temperatures (higher than 70 K,  $70 k_B T \approx$  the localization energy  $E_{\text{loc}}$  of neutral Si donor). In contrast to this behavior, for the AlN single-layered buffer [[layer]] sample, the  $\text{D}^0\text{X}$  peak can be followed up to the room temperature. This observation is consistent with the SIMS results; i.e., the GaN film grown on the single-crystal AlN/ $\text{Si}_3\text{N}_4$ /Si(111) contains much less Si impurities.

**Please replace the paragraph beginning on page 16, line 24 and ending on page 17, line 7, with the following amended paragraph:**

FIG. 4B presents the Arrhenius plots of the luminescence intensities of FX in GaN grown on the single-crystal ~~double-buffer-layer~~ double-layered buffer and D<sup>0</sup>X in GaN grown on the single-layered buffer ~~[[layer]]~~. From the Arrhenius plots, the activation energy of FX ( $E_x$ ) in the GaN grown on the single-crystal AlN/Si<sub>3</sub>N<sub>4</sub>/Si(111) was obtained by fitting the thermal activation relation ~~[[is]]~~ to be about 25 meV, in good agreement with the reported value for FX in undoped GaN. Furthermore, the activation energies of the non-radiative recombination of D<sup>0</sup>X in GaN grown on the single-crystal ~~AlN/Si<sub>3</sub>N<sub>4</sub>/Si(111)~~ AlN/Si(111) can be fitted by using two thermal activation energies ( $E_{a1}$  and  $E_{a2}$ ). The obtained values of  $E_{a1}$  and  $E_{a2}$  correspond well to the known localization energy ( $E_{loc}$ ~6 meV) and donor binding energy ( $E_D$ ~29 meV) of Si impurities in GaN.

**Please replace the paragraph on page 17, lines 11-28, with the following amended paragraph:**

The present invention performed Raman scattering measurements to compare the crystal qualities of GaN epitaxial layers grown on Si (111) by using two different ~~buffer layer systems~~. FIG. 5 displays typical non-polarized Raman spectra in logarithmic intensity scale collected in backscattering geometry along the GaN  $c$  ([0001]) axis (along the growth direction) using the 514.5 nm radiation of an Ar<sup>+</sup> ion laser as a light source, including the dominant phonon peak from Si substrate near 520 cm<sup>-1</sup>. The phonon bands near 568 cm<sup>-1</sup> in each Raman ~~spectrum~~ spectra correspond to the ~~that are~~ GaN E<sub>2</sub> bands. Besides, the A<sub>1</sub> (LO) band near 735 cm<sup>-1</sup> is observed only in the GaN film grown on the single-crystal ~~double-buffer-layer~~ double-layered buffer



and it represents that this GaN film has a lower carrier concentration. According to the previous investigation, the ratio of  $E_2$  to  $A_1(LO)$  ~~to  $E_2$~~  Raman intensity is  $\sim 3$  for the undoped GaN films. The present invention measures the  $E_2$  to  $A_1(LO)$   ~~$A_1(LO)$  to  $E_2$~~  intensity ratio of the GaN film grown on the single-crystal ~~double-buffer layer~~ double-layered buffer is about 3.3 (only slightly larger than 3), indicating the carrier concentration is ~~nearly as low as an undoped GaN film~~ quite low. This is consistent with the SIMS spectra, which show a lower Si concentration in the GaN film grown on Si(111) using a single-crystal AlN/Si<sub>3</sub>N<sub>4</sub> ~~double-buffer layer~~ double-layered buffer.